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THE PROXIMITY EFFECT BETWEEN SUPERCONDUCTING AND NORMAL THIN FILMS IN ZERO FIELD

JOHN CLARKE

Cavendish Laboratory, Cambridge, England

Résumé. — Cette communication passe en revue les expériences sur l'effet de proximité entre une couche mince supraconductrice et une normale, et en donne une bibliographie assez complète. On y discute aussi quelques idées élémentaires sur la théorie de l'effet de proximité. On signale les difficultés particulières rencontrées dans la préparation de spécimens à effet de proximité.

Les expériences sont réparties en trois séries :

I) La température de transition de couches supraconductrices et normales superposées.

II) Les systèmes supraconducteur-normal-supraconducteur capables de supporter une intensité de courant non dissipative.

III) Expériences de tunnel sur les couches doubles.

On compare pour l'ensemble la théorie et l'expérience. L'accord est bon pour I) et II), mais pour III), aussi bien l'interprétation des résultats d'expériences que le calcul théorique des densités d'états sont sensiblement moins satisfaisants.

On discute une tentative pour utiliser l'effet de proximité à l'estimation du paramètre d'interaction de supraconductivité dans les métaux où il est très petit, comme le cuivre : on constate qu'elle est critiquable.

Abstract. — The present paper is a review of experiments on the proximity effect between superconducting and normal thin films and contains a rather complete bibliography. Some elementary ideas of the theory of the proximity effect are also discussed. An account is given of the special difficulties encountered in the preparation of proximity effect specimens.

The experiments are divided into three classes for the purpose of review : (i) The transition temperature of superconducting-normal superimposed films, (ii) Superconducting-normal-superconducting systems which are able to sustain a non-dissipative current flow, (iii) Tunnelling experiments on binary layers. A comparison of theory and experiment is made throughout. It is concluded that for (i) and (ii) there is fair agreement but that for (iii), both the interpretation of the experimental data and the theoretical calculations of the densities of states are somewhat less satisfactory.

Attempts to use proximity effect data to estimate the superconducting interaction parameter in metals where it is very small, such as copper, are discussed and shown to be open to criticism.

I. Introduction. — If a normal metal (N) is in good electrical contact with a superconductor (S), Cooper pairs are able to spread into the normal metal which in some sense then becomes superconducting. Conversely, the « normal » electrons spread into the superconductor, tending to make it « weaker » near the contact. This phenomenon which has become known as the « proximity effect » has been extensively studied both experimentally and theoretically over the past few years. The present paper reviews the various types of experiment which demonstrate the effect and indicates to what extent the theory and experiment are in agreement. No attempt has been made to survey all of the work which has been undertaken in the field but instead a comparatively small number of representative experiments have been selected and their results discussed in some detail. Nor is it the purpose of the article to discuss at length the various theories con-

cerned with the proximity effect : their results will be merely quoted and reference made to the original work.

There are three kinds of experiments which will be discussed in sections IV, V and VI respectively :

(i) **THE TRANSITION TEMPERATURE OF NS LAYERS.** — The transition temperature of a thin superconducting film deposited on a normal metal may be significantly lower than that of the bulk superconductor or indeed, may go to zero. This demonstrates the « quenching » of superconductivity by the proximity of the normal metal.

(ii) **CRITICAL CURRENTS OF SNS SANDWICHES.** — If superconductivity does extend into the normal layer, it should be possible to pass a supercurrent through a normal metal sandwiched between two superconductors, provided that it is not too thick.

(iii) **TUNNELLING EXPERIMENTS ON NS LAYERS.** — Numerous experiments have been performed in which

an oxide layer is grown on to one side of the sandwich and covered with a third metallic layer. The I-V characteristic of this tunnel junction enables one to study the electronic density of states in the sandwich and to show, for example, that some degree of superconductivity extends well into the normal metal.

Inevitably, some methods have been omitted from this list [63, 64]. Of these, the most important is probably the microwave surface impedance technique used by Fanelli and Meissner [49] and by Waldram [68].

Before embarking upon a description of the experiments themselves, we shall discuss the considerable difficulties encountered in the preparation of proximity effect specimens (Section II). This will be followed by a section dealing with those elements of the theory necessary to gain some insight into the experimental results.

II. Preparation of proximity effect specimens. —

At a sufficiently large distance from the NS interface the electronic behaviour in each metal must become independent of the presence of the other. Consequently in order to observe proximity effects it is necessary to prepare specimens in which at least one and often both metals are « thin ». Thin film systems are prepared by evaporation or sputtering in vacuo or by electrolytic deposition. In all cases, it is necessary to take elaborate precautions to avoid the possibility of spurious effects.

The first important consideration is the choice of materials. Chiou and Klokholm [3] discovered that a system of silver and tin showed anomalies in its transition temperature because of the formation of Ag_3Sn , rather than by means of a simple effect between two pure films. It is therefore essential to select two metals which do not form intermetallic compounds.

Rose-Innes and Serin [16] demonstrated that the interdiffusion of the two metals can also considerably affect the experimental results, because of the drastic effects on the mean free path near the boundary resulting from the impurity introduced. It is thus necessary to choose two metals whose mutual solubility is as low as possible, particularly in view of the fact that diffusion rates in vacuum-deposited thin films may be one or two orders of magnitude greater than in bulk materials [3]. Several authors [for example, 6, 9] have overcome this difficulty by depositing their films on to substrates maintained at low temperatures, where diffusion rates become much lower. In this case, it is imperative that the films be maintained at the low temperature and not exposed to the atmosphere before the experiments have been performed. The low temperature deposition results in a high defect density and Bergmann et al. [2] showed that this leads to a greatly

enhanced diffusion rate at a given temperature, so that warming up the film to room temperature can have disastrous consequences. In addition, the warming up of a specimen will introduce annealing effects which may be very difficult to estimate. Hauser et al. [8] have pointed out that exposure to moist oxygen at room temperature may set up an electrochemical cell reaction which oxidizes the more anodic metal at the interface. This results in a decoupling of the two metals and again proceeds very rapidly in systems deposited at low temperatures. Miles and Smith [13] discovered a mechanism in Al/In specimens by which hydrogen was introduced at the interface, again tending to decouple the two films. When the specimen was exposed to the atmosphere, water-vapour oxidized the top surface of aluminium and released atomic hydrogen. Some of the hydrogen diffused into the film and recombined to form molecular hydrogen which was trapped at the interface. Similar effects were noted in other systems.

It is apparent that if the specimen is to be made at low temperatures, then only an *in situ* preparation is acceptable. However, Bassewitz and Minnigerode [36] have demonstrated that copper deposited at 77 °K has a density of about two-thirds the bulk value. Consequently the results of experiments on films deposited and kept at low temperatures, although they may provide excellent qualitative evidence for the proximity effect, must be regarded with some suspicion as far as the superconducting properties of the bulk normal metal are concerned. However, anomalous densities are not observed in all materials and clearly a systematic investigation is urgently needed. One may thus conclude that there is some justification for preparing specimens at room temperature where the density is more nearly that of the bulk material, provided that the mutual solubilities are sufficiently low. This problem is greatly alleviated by deliberately adding a suitable impurity to each metal: the resulting short mean free path minimizes the effects of diffusion near the boundary. The effects of short exposure of these specimens to the atmosphere are questionable, but one might hope that by careful choice of materials the effects may not be too serious. There are in fact remarkably few pairs of metals which may be regarded as satisfactory for proximity effect measurements: examples are Pb/Cu, Pb/Ag, Pb/Fe and In/Zn.

A number of workers [6, 8, 10] have reported that the transition temperature of a binary layer may be dependent upon the order in which the materials are deposited. There are at least two possible mechanisms for this effect. An oxide barrier may grow on the first metal before the second is deposited on top of it. The

oxide growth rate on (say) lead is much greater than on (say) copper, so that this would lead to the observed irreversibility. The effect may be minimized by evaporation in an ultra-high vacuum system or by the use of getter-sputtering techniques [66]. It is thought that an oxide barrier was responsible for the absence of a proximity effect in the Al/Sn system studied by van Gorp [21]. The second cause of irreversibility is the dependence of the morphology of a thin film on the nature of the substrate [8]. Thus lead deposited on copper may nucleate in a different way from lead deposited on glass and the resultant variations in mean free path and density near the interface will obviously influence the proximity effect. The use of alloys which are so dirty that impurity scattering dominates the resistivity may effect a partial cure of this problem.

It is clear that experimental specimens are very far removed from the parallel-sided homogeneous models of the theory. In particular, the character of the interface has been completely ignored experimentally, but it seems likely that there will be considerable distortion of the material in this region which will modify the proximity effect appreciably. This topic has been discussed at length by van der Merwe [67]. Much remains to be achieved in specimen preparation before experimental results can be compared with some of the finer points of the theory.

III. Theory. — The B. C. S. theory of superconductivity [35] introduces an interaction parameter V which represents the effective electronic interaction. The condition for superconductivity is $V > 0$. A knowledge of V for a particular metal gives us its energy gap at absolute zero $\Delta(0)$ and its transition temperature T_C through the relation :

$$2 \Delta(0) = 3.52 k_B T_C = 4 \hbar \omega_q \exp(-1/NV), \quad (1)$$

where $N(0)$ is the density of states at the Fermi surface and ω_q an average phonon cut-off frequency. Morel and Anderson [58] have pointed out that V can be regarded as a point interaction. Thus in a system of two different superposed metals, we can suppose that the interaction parameter changes instantaneously at the boundary. It is the recognition of the fact that this is *not* true for the electronic wave function which leads us to the proximity effect. The fact that the wave function cannot fall abruptly to zero is implied by the phenomenological theory of Ginzburg and Landau [52] who show that the superconducting order parameter cannot vary markedly within a distance known as the Ginzburg-Landau coherence length. Unfortunately, as we shall see, the self-consistency equation obeyed by

the order parameter is non-local in most proximity effect situations so that the approximation represented by the Ginzburg-Landau theory is not always valid. We shall therefore briefly describe the conclusions of the present microscopic theory which has proved quite successful in explaining the experimental results.

The phenomena of superconductivity are associated with the presence of a certain type of ordering in the electronic wave functions in the metal. In the B. C. S. theory the ordering is thought of as a correlation in the occupation of the one-electron states at \mathbf{k} and $-\mathbf{k}$ in reciprocal space ; the degree of order is expressed by a *gap parameter*, Δ_k , which is a function of momentum. This point of view is appropriate only when the degree of order is uniform in space : Δ_k at the Fermi surface is then just Δ , the energy gap. Where there is an inhomogeneous situation, such as in the proximity effect, a different formalism is required. The degree of order becomes a function of position and may be expressed as a *condensation amplitude*, $F(\mathbf{r}) = \langle \psi_{\uparrow}(\mathbf{r}) \psi_{\downarrow}(\mathbf{r}) \rangle$, where $\psi_{\uparrow}(\mathbf{r})$ is an electron annihilation operator (see, for example [55]). $|F(\mathbf{r})|^2$ is essentially the probability of finding a Cooper pair at \mathbf{r} , that is, it represents the superfluid density. In this notation it is the *pair potential*, $\Delta(\mathbf{r}) = V(\mathbf{r}) F(\mathbf{r})$ which is the analogue of Δ in the B. C. S. theory.

The de Gennes-Guyon theory of the proximity effect [40, 42] uses the Gor'kov self-consistent integral equation which is valid for small $\Delta(\mathbf{r})$:

$$\Delta(\mathbf{r}) = \int d^3 \mathbf{r}' K(\mathbf{r}, \mathbf{r}', T) \Delta(\mathbf{r}'), \quad (2)$$

where K is the kernel with a range (coherence length) :

$$\xi(T) = \left(\frac{\hbar v_F l}{6 \pi k_B T} \right)^{1/2}. \quad (3)$$

v_F is the Fermi velocity and l the electronic mean free path. Equation (2) expresses the cooperative nature of the effect (just as the B. C. S. integral equation does) in that pairing at a point \mathbf{r} is related to that at all other points \mathbf{r}' . The theory is valid strictly only for dirty systems ($l \ll \xi$) and near the transition temperature of the NS sandwich (T_{CNS}).

De Gennes [40] considers specifically a NS sandwich (see Fig. 1) in which (2) becomes a one-dimensional equation and shows that the boundary conditions on $\Delta(x)$ at the interface are :

$$\frac{\Delta_N(0)}{N_N V_N} = \frac{\Delta_S(0)}{N_S V_S} \quad (4a)$$

and

$$\frac{\xi_N^2}{V_N} \left. \frac{d\Delta_N}{dx} \right|_0 = \frac{\xi_S^2}{V_S} \left. \frac{d\Delta_S}{dx} \right|_0. \quad (4b)$$

Equation (4a) shows that in general $\Delta(x)$ is discontinuous at the boundary but that F/N is continuous: this implies that the fraction of electrons paired is conserved across the interface.

De Gennes uses (2) to derive the shape of $\Delta(x)$ across a NS sandwich in which each film is thicker than a coherence length (see Fig. 1). In the superconductor,

$$\Delta_S(x) \propto \frac{\cos k_S(x + d_S)}{\cos k_S d_S}, \quad (5)$$

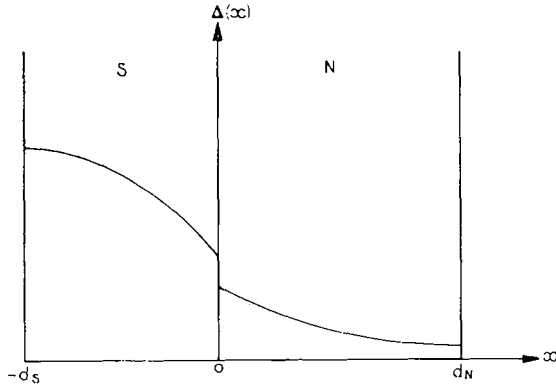


FIG. 1. — Variation of the pair potential $\Delta(x)$ across a NS sandwich for $V_N > 0$. In general, there is a discontinuity in $\Delta(x)$ at the interface.

where

$$k_S^{-1} = \left(\frac{\hbar v_{FS} l_S}{6 \pi k_B T} \right)^{1/2} \left(\frac{2}{\ln(T_{CS}/T)} - 1 \right)^{1/2}. \quad (6)$$

k_S^{-1} is essentially the Ginzburg-Landau coherence length [52] which varies as $(T_{CS} - T)^{-1/2}$ near the transition temperature of the superconductor, T_{CS} . In the normal metal, far from the interface (the « one frequency approximation », $k_N x \gg 1$), $\Delta(x)$ has an exponential form :

$$\Delta_N(x) \propto \exp(-k_N x), \quad (7)$$

where

$$k_N^{-1} = \left(\frac{\hbar v_{FN} l_N}{6 \pi k_B T} \right)^{1/2} \left(1 + \frac{2}{\ln(T/T_{CN})} \right)^{1/2} \quad (8)$$

represents the depth of penetration of pairs into the normal metal. It should be emphasized that this last result is *not* true for $x \lesssim k_N^{-1}$ in which region the contribution of the superconductor to the kernel in (2) is substantial. Notice that for the special case $V_N = 0$, k_N^{-1} reduces to ξ_N and $\Delta_N(x)$ is everywhere zero in the normal metal (from (4a)). In the region of interest, $T_{CN} < T < T_{CS}$, k_N and k_S have opposite temperature dependences.

Equation (8) is of interest in that it contains T_{CN} . Thus proximity effect experiments which measure k_N^{-1} on a system where the normal metal is a superconductor

above its transition temperature provide a useful test of the theory. Conversely, experiments using a normal metal which is not a known superconductor are a means of determining T_{CN} from (8) and thus $N_N V_N$ through (1). Unfortunately, the theory is valid only for $V_N \geq 0$; for $V_N < 0$, it is necessary to take into account retardation effects. A recent preliminary calculation by de Gennes [46] indicates that $k_N^{-1} = \xi_N$ for $V_N \leq 0$, so that the cases $V_N = 0$ and $V_N < 0$ are indistinguishable. Nevertheless, such measurements are of some interest, particularly in view of the prediction of Morel and Anderson [58] that V_N is always positive so that all metals (except those which are magnetic) should become superconducting at a sufficiently low temperature. However, as NV is decreased below about 0.1, the corresponding transition temperature drops off exceedingly rapidly to values which cannot be observed directly. The proximity effect is therefore a useful means of measuring NV in metals where it is small, provided that it is positive. In the description of experiments which follows, the values of NV in copper obtained by various authors will be quoted but in the light of the recent theoretical development, it now seems likely that negative values (usually the limits of experimental uncertainty) should be disregarded.

IV. The transition temperature of NS layers. —

(i) NON-MAGNETIC NORMAL METAL. Measurements of the transition temperature of NS layers have been the most popular type of proximity effect experiment [1-21]. The work of Hilsch [9] on Pb/Cu was the first to include data on the mean free paths in the films and we shall consider his results in some detail. His films were deposited at 10 °K and not warmed up or exposed to oxygen until after the experiment. This means that the results represent a genuine proximity effect but presumably the copper films had a low density which casts some doubt on the quantitative deductions. The mean free paths were certainly short enough to ensure that the films were in the dirty limit.

The experimental results of Hilsch et al. [11] are reproduced in figures 2 and 3. In figure 2, the transition temperature of the sandwich is plotted as a function of copper thickness for a number of thicknesses of lead. These results can be readily interpreted qualitatively. For the 100 Å lead film, an increase in the thickness of copper initially lowers the transition temperature considerably. When the film thicknesses are much less than their respective coherence lengths, each Cooper pair extends over the entire system and is subject to an average value of NV which is smaller than its value in the isolated lead film [39]. As the copper thickness is increased from zero, this average

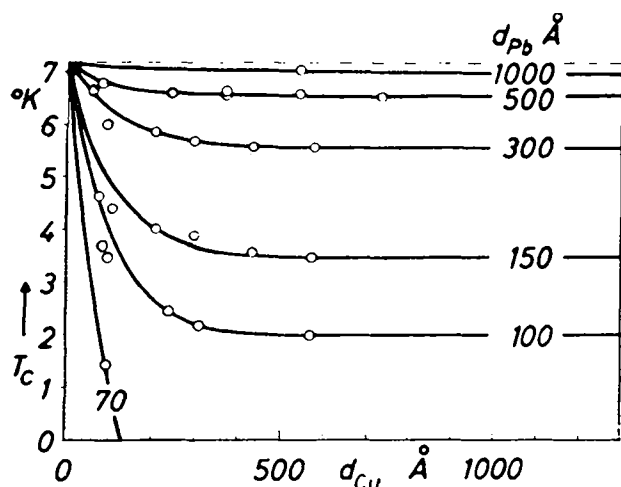


FIG. 2. — Transition temperature (T_c) of Pb/Cu sandwiches as a function of Cu thickness (d_{Cu}) for various thicknesses of Pb. (Reproduced from Hilsch et al. [11].)

value will decrease and T_{CNS} will be sharply depressed, in view of its exponential dependence on NV . However, once the copper has a thickness of the order of a few coherence lengths (in this case about 200 Å) the addition of more copper can have no effect upon the lead and T_{CNS} will tend to a limiting value depending on the thickness of the superconductor. Obviously, the thinner the lead, the greater will be the effect of a given thickness of copper, giving rise to the vertical displacement of the curves with varying thicknesses of lead. This is also illustrated in figure 3, where the transition temperature is plotted against the thickness of lead films deposited on a thick copper film ($\gg \xi_N$) for two different values of mean free path in the latter.

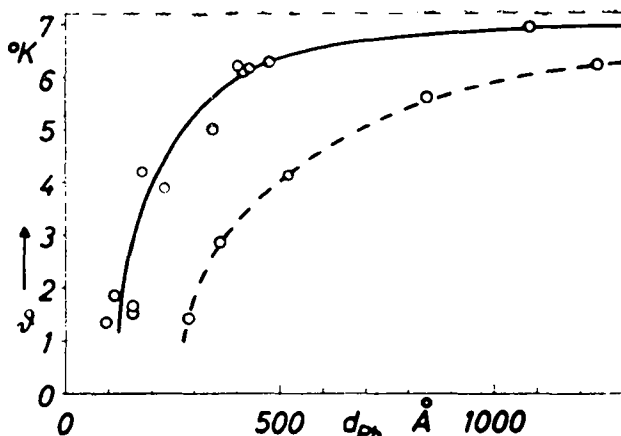


FIG. 3. — Transition temperature (t) of Pb films on a thick Cu film as a function of Pb thickness (d_{Pb}). Mean free path in Pb 55 Å, in Cu 40 Å (solid curve) and 800 Å (dotted curve). (Reproduced from Hilsch et al. [11].)

Since the copper can only affect the lead over a distance of a few coherence lengths, those specimens with thick layers of lead will have a transition temperature identical with the bulk value. As the thickness is reduced, the copper has an increasing effect on the transition temperature, eventually reducing it to below an observable value. The dotted curve shows the effect of increasing the mean free path of the copper by annealing it prior to the lead deposition. This increases the coherence length of the copper which therefore depresses the transition temperature still more for a given thickness of lead. By this and a similar experiment in which the mean free path of the lead was varied, Hilsch et al. [11] were able to show that the coherence length was proportional to $l_N^{1/2}$, in accordance with (8).

Werthamer [69] has used the de Gennes-Guyon theory to derive the following relations between T_{CS} , T_{CN} and T_{CNS} :

$$\chi(\xi_S^2 k_S^2) = \ln(T_{CS}/T_{CNS}), \quad (9)$$

$$\chi(-\xi_N^2 k_N^2) = \ln(T_{CN}/T_{CNS}), \quad (10)$$

$$N_S \xi_S^2 k_S \tan(k_S d_S) = N_N \xi_N^2 k_N \tanh(k_N d_N), \quad (11)$$

where $\chi(Z) = \Psi(Z/2 + 1/2) - \Psi(1/2)$

and $\Psi(Z) = \Gamma'(Z)/\Gamma(Z)$

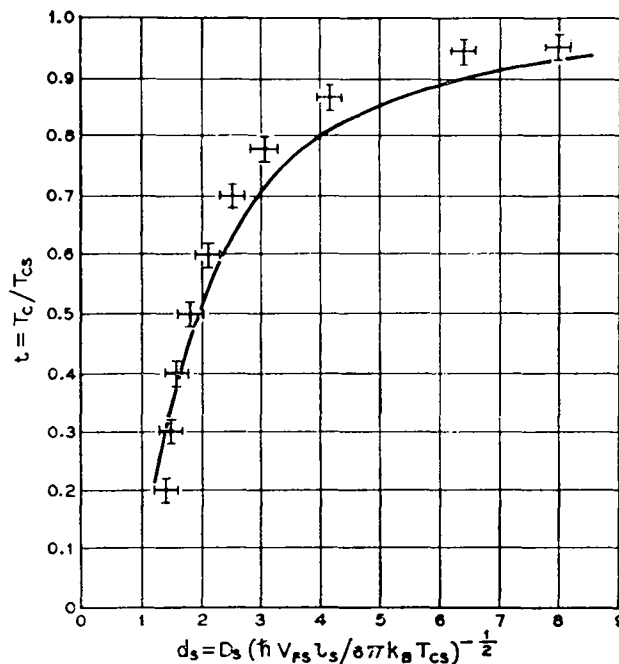


FIG. 4. — Reduced transition temperature (t) of Pb/Cu sandwiches as a function of reduced lead thickness (d_s) as predicted by the Werthamer theory (solid curve). Data points are from Hilsch's data [9, 11]. (Reproduced from Werthamer [69].)

is the digamma function. In figure 4 (taken from [69]) the curve is the reduced transition temperature as a function of the reduced thickness of lead (i. e. in units of a coherence length) calculated from (9)-(11) and the points are from the experimental results of Hilsch [9, 11]. The agreement must be considered quite good, bearing in mind the experimental errors. De Gennes [40] has used Hilsch's results to estimate $(NV)_{Cu}$ as + 0.05 with possible limits of - 0.06 and + 0.10, although no allowance has been made for the anomalous density of the copper resulting from the low temperature deposition.

Hauser and Theuerer [5] have performed similar experiments in which the copper was replaced by a thick film of aluminium, which has a transition temperature of about 1.2 °K. The transition temperature of the sandwich as a function of lead thickness is plotted in figure 5. As the thickness of lead is lowered, the transition temperature falls off as before, but flattens out at low values towards the transition temperature of aluminium, as we should expect. The solid line is

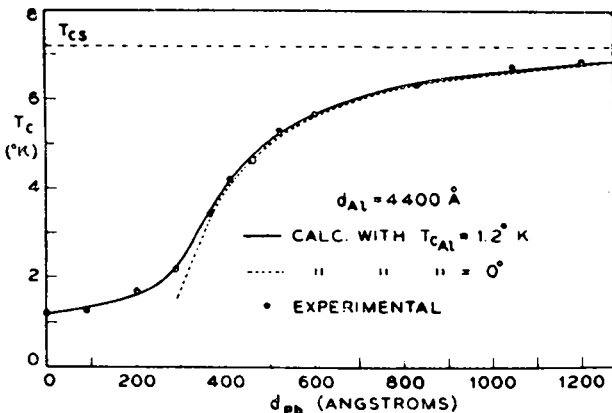


FIG. 5. — Transition temperature (T_C) of Pb/Al sandwiches for a constant Al thickness of 4400 Å as a function of the Pb film thickness (d_{Pb}) (Reproduced from Hauser and Theuerer [5].)

calculated from the de Gennes-Guyon-Werthamer theory and agrees very closely with the experimental results with $T_{CN} = 1.2$ °K (solid line). However, the dotted line represents the theoretical curve for $T_{CN} = 0$ °K and we see that it diverges significantly from the other only at low transition temperatures. The transition temperature of aluminium could be determined with reasonable accuracy only below about 4 °K. This insensitivity of the theory to the value of $N_N V_N$ well above T_{CN} will be discussed again later.

(ii) **MAGNETIC NORMAL METAL.** — It is well known that small quantities of magnetic impurity in a super-

conductor may suppress its transition temperature enormously. One might therefore expect a layer of magnetic atoms to profoundly affect the properties of a superconductor adjacent to it. Hauser et al. [7, 8] have investigated the transition temperatures of binary layers in which the normal metal was a ferromagnetic, an antiferromagnetic or a dilute magnetic alloy. Figure 6 shows the transition temperature of lead films of various thicknesses deposited (at 77 °K) on a 1000 Å film of iron. The effect of the iron film is qualitatively similar to that of a non-magnetic metal but is much stronger. Hauser et al. first try to explain their results by putting $T_{CN} = 0$ °K in equation (10) to obtain the solid curve of figure 6. It is clear that this does not fit the experimental points. The assumption that the interaction in the iron is infinitely repulsive gives rise to the dashed curve which is very little different from the previous curve and still disagrees

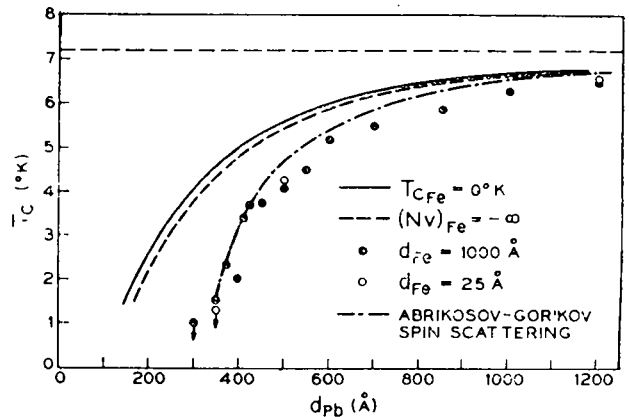


FIG. 6. — Transition temperature (T_C) of Pb/Fe sandwiches as a function of Pb thickness (d_{Pb}). (Reproduced from Hauser et al. [8].)

with the results. Obviously, the theory must be modified to account for the ferromagnetic interaction.

Abrikosov and Gor'kov [34] and later de Gennes and Sarma [45, 62] have formulated theories to account for the effect of a low concentration of magnetic impurities on the properties of superconductors. Such impurities greatly reduce the transition temperature because the two mates ($\mathbf{k}_1, \mathbf{k}_1$) of a Cooper pair see vastly different exchange potentials. This depairing effect depresses the transition temperature to a value T'_{Cs} , where

$$\ln(T_{Cs}/T'_{Cs}) = \chi(\hbar/2 \pi \tau_s k_B T'_{Cs}). \quad (12)$$

τ_s is the lifetime of a Bloch state of the Fermi surface due to the interaction between the impurities and the conduction electrons. There is obviously a close ana-

logy between equations (12) and (10). Hauser et al. assume that a modified version of (10) can be used to interpret their results and replace it by

$$\ln(T_{CN}/T_{CNS}) = \chi(-\xi_N^2 k_N^2 + \hbar/2 \pi \tau_S k_B T_{CNS}). \quad (13)$$

They choose $T_{CN} = 0$ and by fitting the theory at one experimental point find $\tau_S \approx 10^{-14}$ s, corresponding to a scattering length in the iron of about 100 Å. Equations (9), (11) and (13) then produce the lowest curve of figure 6, which is in fair agreement with the data.

The pair penetration depth in the iron may be estimated from equation (13) to be about 6 Å. This means that only a very thin layer of iron is necessary to produce the observed depression in transition temperature: results obtained with a 25 Å layer of iron were indistinguishable from the previous ones, as is shown in figure 6. The main conclusion of these results is that one can write the boundary condition $\Delta(\mathbf{r}) = 0$ at an interface with a ferromagnetic material. This is confirmed by the experiments of Groff and Parks [4] and the calculations of Fulde and Maki [51].

Hauser et al. performed similar experiments on Pb/Cr, Cr being an anti-ferromagnet, and were able to obtain a reasonable fit using equation (13). Probably the grain size was sufficiently small for the random impurity spin description to remain valid. In a final set of experiments they used a dilute magnetic alloy of Pb containing 2.9 % Gd as the normal metal, its transition temperature being 1.52 °K. τ_S was obtained independently by measurements on a separate alloy film. The proximity effect results were in good agreement with the theoretical predictions of equation (13).

V. Critical currents in SNS sandwiches. — Consider a normal metal sandwiched between two superconductors. Provided that the thickness of the middle layer is not too great, the superfluid density will be everywhere non-zero and one might expect to be able to pass a supercurrent between the superconductors through the normal metal. The system is in many ways analogous to a Josephson junction [54] in which Cooper pairs tunnel through a thin insulating barrier separating two superconductors (SIS) although, as we shall see, there are some important differences.

The first experiments on this system were those of Meissner [22], who measured the I-V characteristics of crossed copper-coated tin wires. He found that the resistance between the wires for small currents did indeed vanish for sufficiently low thicknesses of copper. However, because of the uncertainty in the contact area these experiments are difficult to interpret quantitatively.

The present author has recently studied Pb-Cu-Pb sandwiches in some detail. The junctions were made by evaporating successively on to a water-cooled glass substrate a strip of lead, a circular layer of a copper-aluminium alloy and a second cross-strip of lead (Fig. 7). The evaporation was performed in a vacuum of about 10^{-8} torr, so that oxidation of the specimen during preparation was minimized. The mean free path in the alloy was estimated, from the resistance of a strip evaporated simultaneously on to the same slide, to be typically 100 Å so that the normal metal was in the dirty limit. The specimen area defined by the overlap of the lead strips was about 5×10^{-4} cm². The four terminal I-V characteristic was measured by passing a current through the specimen and measuring the voltage developed by means of a voltmeter with a sensitivity of 10^{-14} volts [38].

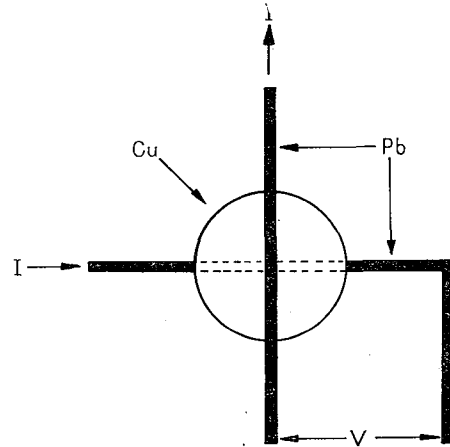


FIG. 7. — Configuration of Pb/Cu/Pb sandwich.

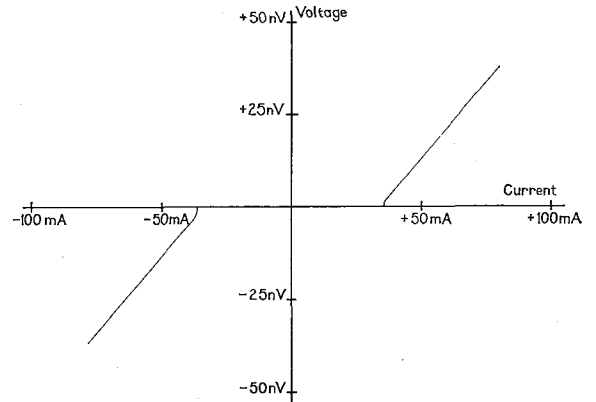


FIG. 8. — I-V characteristic for a typical Pb/Cu/Pb sandwich.

A typical I-V plot is shown in figure 8. We see that (within the sensitivity of the voltmeter) no voltage

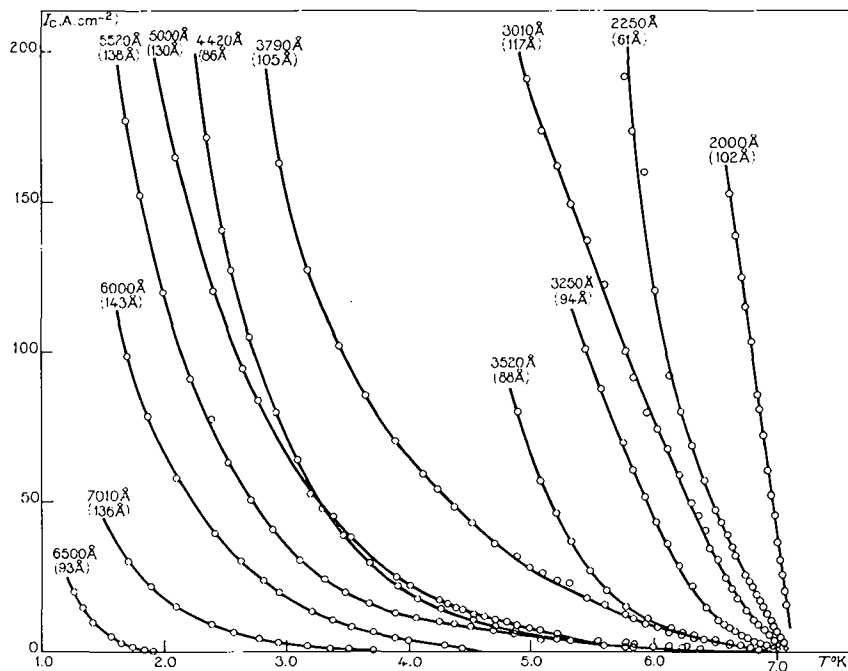


FIG. 9. — Critical current density (I_C) as a function of temperature (T) for various thicknesses of Cu in Pb/Cu/Pb sandwiches. The numbers in brackets refer to the mean free path of the Cu.

appears across the sandwich until the current exceeds a well-defined critical value, I_C . For currents greater than this, the junction exhibits a resistive behaviour, with which we are not concerned here. Notice however, that the voltages developed are very much smaller than those involved in SIS junctions: this is a reflection of the very low resistance of the normal metal, about 10^{-6} ohms, compared with a value of perhaps 1 ohm for the insulator. It is also interesting to note the relatively thick copper layers through which a supercurrent may be passed, a few thousand Angstroms as opposed to 10-20 Å of insulator.

Most of the experiments were directed at the measurement of critical current (I_C) as a function of temperature (T), normal metal thickness ($2a$) and normal metal mean free path (l_N). Figure 9 is a graph of I_C against T for varying thicknesses of copper with (essentially) constant mean free path, each plot being an average of the results from two specimens. The very rapid increase in I_C as T is reduced is striking and in complete contrast to the case of SIS junctions where the curve flattens out at lower temperatures. At a given temperature, the critical current is monotonic in thickness of copper, except for the 6,500 Å and 7,010 Å samples, which have been reversed by the

abnormally large difference in the mean free path of copper. Figure 10 shows the thickness dependence of

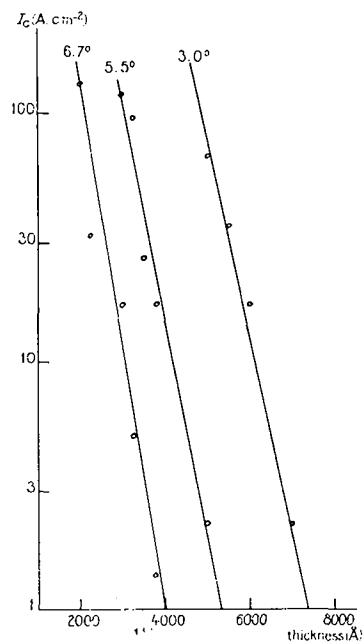


FIG. 10. — Critical current density (I_C) against Cu thickness in Pb/Cu/Pb sandwiches at three different temperatures.

the critical current at three different temperatures. As we might expect, I_C varies as $\exp(-\text{const. } 2a)$.

Using the one-frequency approximation ($k_N a \gg 1$) which is adequate in these experiments, de Gennes [41, 42] has shown that the critical current of an SNS sandwich is given by

$$I_C = Ak_S^2 k_N^{-1} \Delta_{Pb}^2 \exp(-2 k_N a). \quad (14)$$

A is a constant and $\Delta_{Pb}(T)$ the energy gap in the lead far from the copper insert. The result is strictly true near T_{CNS} and for weak-coupling [54], that is, in the situation where the current flowing is so small that it affects only the phase and not the amplitude of the energy gap parameter. The exponential dependance is as we should expect; the term $k_S^2 k_N^{-1}$ expresses the depression of the gap parameter to below the bulk value at the lead-copper interface. Near $T_{CS} (= T_{CNS}$ if the lead is very thick), both Δ_{Pb} and k_S vary as $(1-t)^{1/2}$, where $t = T/T_{CS}$, so that

$$I_C \propto (1-t)^2. \quad (15)$$

This result is an important test of the de Gennes theory. It should be contrasted with the SIS junction for which $I_C \propto (1-t)$; the order parameter in the superconductor is *not* depressed by the proximity of an insulator and only the temperature dependence of Δ_{Pb} is involved. Figure 11 shows the dependance of

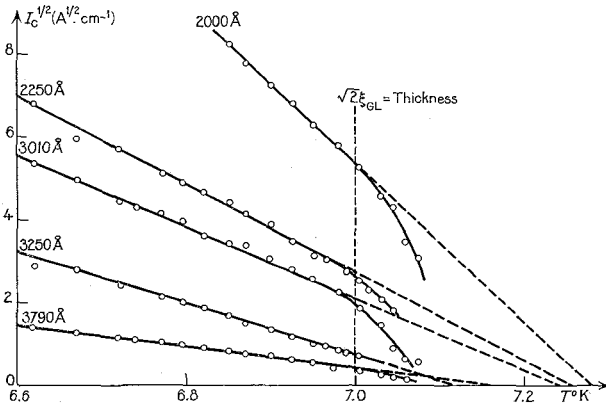


FIG. 11. — $I_C^{1/2}$ against temperature (T) for five thicknesses of Cu in Pb/Cu/Pb sandwiches

$I_C^{1/2}$ upon T near T_{CNS} for five thicknesses of copper. We see that the slope is sensibly constant between 6.6 °K and 7.0 °K: this may be taken as a convincing verification of equation (15). As T is increased above about 7.0 °K, I_C falls off more rapidly because the coherence length in the lead, which is proportional to $(1-t)^{-1/2}$, becomes comparable with the thickness

of the lead strips (7 000 Å), so that the energy gap is depressed throughout the superconductor.

The behaviour of I_C well below T_{CNS} is difficult to interpret quantitatively. However, at low temperatures the term $\exp(-\text{const. } T^{1/2})$ is dominant in equation (14): this would qualitatively explain the very rapid increase in I_C as the temperature is lowered.

A separate series of experiments has shown that $I_C \propto \exp(-\text{const. } T_N^{1/2})$ as predicted by (14). The thickness dependence of I_C (Fig. 10) provides an estimate of $(NV)_{Cu}$. Inserting the experimental values into equation (8) gives an upper limit on $(NV)_{Cu}$ of 0.11, corresponding to a transition temperature of 6×10^{-2} °K.

VI. Tunnelling experiments on NS layers. — The experiments on the proximity effect described above give information upon the behaviour of the superfluid only and to obtain the excitation spectrum of a NS layer from a transport process it is necessary to use a dissipative process, such as one-electron tunnelling [23-33]. This technique is becoming increasingly important in the investigation of the proximity effect but one should bear in mind that the fabrication of good tunnelling junctions is still a rather difficult and empirical undertaking.

When tunnelling occurs between a normal metal and a superconductor through a thin insulating barrier, the normalized differential conductance at a voltage V is related to the electron density of states in the superconductor by (see, for example [47]):

$$\frac{dI}{dV} = \int_{-\infty}^{\infty} N_S(\epsilon) \frac{\partial}{\partial V} f(\epsilon + eV) d\epsilon. \quad (16)$$

$f(\epsilon)$ is the Fermi function and $N_S(\epsilon)$ the normalized excitation density of states at energy ϵ which for a B. C. S. superconductor is:

$$N_S(\epsilon) = \begin{cases} \frac{|\epsilon|}{\sqrt{\epsilon^2 - \Delta^2}} & \epsilon \geq \Delta, \\ 0 & \epsilon < \Delta. \end{cases} \quad (17)$$

The tunnelling characteristic corresponds to the density of states at the *surface* of the « superconductor » which in a proximity effect experiment may be either side of a NS sandwich. At $T = 0$ °K, $\partial/\partial V[f(\epsilon + eV)]$ becomes a delta function and dI/dV is simply $N_S(\epsilon)$. At a finite temperature, dI/dV becomes a smoothed version of $N_S(\epsilon)$; a deconvolution of the integral gives $N_S(\epsilon)$ but unfortunately, large errors may be introduced by comparatively small discrepancies in dI/dV .

The theoretical calculation of $N_S(\epsilon)$ in a NS sandwich is generally exceedingly difficult and it should be

emphasized that the excitation spectrum in this case is usually quite different from that of a B. C. S. superconductor. However, there are two limiting situations in which the problem has been tackled theoretically although most of the experiments appear to lie somewhere between the two extremes and the distinction between the two kinds of experiment is not always obvious.

The first situation is concerned with films which are clean enough for bulk scattering to be unimportant ($l \sim d$) and sufficiently thin for the order parameter in each metal to be taken as a constant ($d \ll \xi$). The theory of Rowell and McMillan [31] is then applicable, at temperatures well below T_{CNS} , and derives expressions for $N_S(\epsilon)$ on both S and N sides. Unfortunately, there are probably no experiments which really satisfy these rather stringent requirements although those of Hauser [28] and Adkins and Kington [23] show at least a qualitative fit to the theory. A question of particular importance is the value of the minimum excitation energy, Δ_m , likely to be seen in a tunnelling experiment on either side of a NS sandwich. The Rowell-McMillan theory predicts that this minimum value will be the same on each side. The underlying physical reason is presumably that although the lowest lying excitation tends to be localized in the normal metal, it can be seen from the S side provided that the superconductor is not too thick. The estimation of Δ_m from the tunnelling characteristic is by no means straightforward but it may be attempted in one of two ways. At very low temperatures, Δ_m may be taken as the voltage at which dI/dV is unity; however at higher temperatures this is no longer true so that the method is of limited application. Alternatively, we may measure the zero-voltage conductivity $(dI/dV)_0$ which is roughly proportional to $\exp(-\Delta_m/kT)$: values of $(dI/dV)_0$ calculated for a B. C. S. model [37] may then be used to estimate Δ_m . It is clear that neither method is very satisfactory.

The second theoretically tractable situation is that in which the samples are dirty ($l \ll \xi$) and near T_{CNS} . Before discussing it we shall first briefly introduce the concept of *gaplessness* in superconductivity. Reif and Woolf [61] measured the electrical resistance and tunnelling characteristic of superconducting films containing magnetic impurities. As the impurity concentration was increased, they found that the gap decreased twice as rapidly as the transition temperature; an indium film containing 1% iron was resistanceless below 3 °K but displayed no gap. The magnetic ions introduce spin-dependent interactions which shorten the lifetime of the conduction electrons,

broadening the energy levels. At a high enough impurity level, the states above the gap become sufficiently smeared out into the gap for zero-energy excitations to exist [60]. However, in this gapless regime it is still possible for the system to support a zero-frequency supercurrent.

Woolf and Reif [33] also performed experiments on tunnelling into superconductors backed by magnetic films and found that the gap was smeared by an amount which increased as the thickness of the superconductor was decreased. Subsequently, Fulde and Maki [50] proposed that a *non-magnetic* normal film should induce gaplessness into an adjacent superconductor, in a dirty sandwich near T_{CNS} (*). De Gennes and Mauro [43] have criticized [50]. They insist that the density of states always contains a gap (except for the special case, $V_N = 0$) but show that the experimental quantity (dI/dV) may under certain circumstances display a « quasi-gapless » behaviour somewhat similar to what was predicted by Fulde and Maki. De Gennes [40] has also pointed out that if the superconductor is rather thin we should expect to see a minimum excitation energy at the S surface identical to the one in the normal metal, just as in the case of the thin clean sandwich. The problem of calculating $N_S(\epsilon)$ itself has not been solved for the general case.

It is apparent that the interpretation of tunnelling data is difficult; however, a useful idea to have in mind is the fact that $(1 - (dI/dV)_0)$ (which is zero for a normal metal and unity for a superconductor at absolute zero) represents the « degree of superconductivity » present. We shall now briefly describe the experimental results.

(i) THIN ($d \ll \xi$) « CLEAN » ($l \sim d$) FILMS AT LOW TEMPERATURES ($T \ll T_{\text{CNS}}$).

Adkins and Kington [23] measured the tunnelling characteristics of Al/Al₂O₃/Pb/Cu and Al/Al₂O₃/Cu/Pb systems deposited on to a glass substrate at room temperature; the lead was in the clean limit, but the copper was not. Figure 12 shows the characteristic obtained by tunnelling into a pure lead film, into a 330 Å layer of copper on a thick lead film and into a B. C. S. superconductor (calculated) fitted to the latter at $dI/dV = 1$. The important point to notice is that the density of states induced into the copper is much higher than the B. C. S. distribution just above the gap. Adkins and Kington attribute this to localisation of the low energy excitations in the copper. Now in the

(*) Maki and Fulde [56] and Parks [59] have discussed the equivalence of various pair-breaking mechanisms in superconductors.

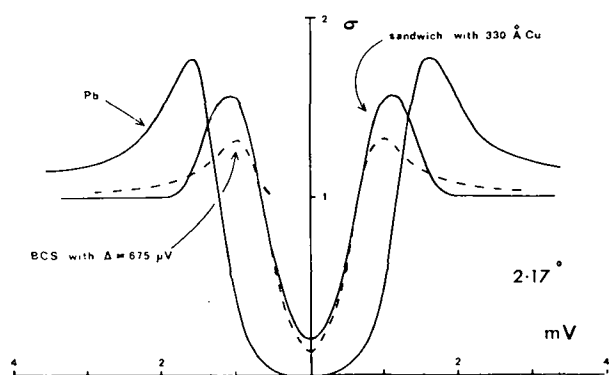


FIG. 12. — Comparison of characteristics for tunnelling into pure lead, into the copper side of a Pb/Cu sandwich (both experimental) and into a B. C. S. superconductor fitted at the voltage for which $dI/dV = 1$ (theoretical). (Reproduced from Adkins and Kington [23].)

B. C. S. formulation, the energy of an excitation is $E_k = (\epsilon_k^2 + \Delta_k^2)$. If we assume that a similar result holds for the NS sandwich but that $\Delta(x)$ decreases for increasing distances into the copper, we see that it is energetically favourable for the low energy excitations to exist in the copper rather than the lead. This was confirmed by tunnelling into a thin lead film deposited on copper: the suppression of the states just above the gap was clearly demonstrated.

The value of Δ_m at the copper surface (deduced from $(dI/dV)_0$) as a function of copper thickness is shown in figure 13. Notice that Δ_m decreases with increasing

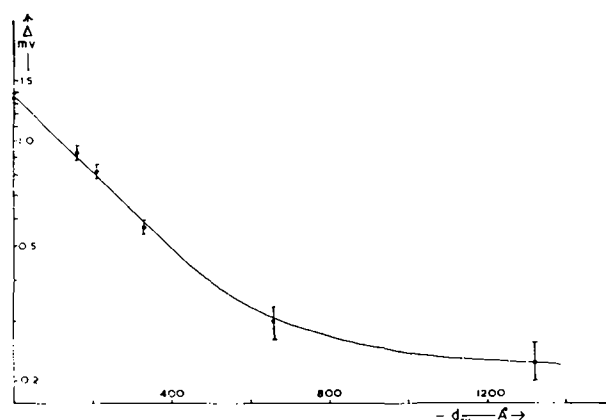


FIG. 13. — The energy gap (Δ_m) at the copper surface in a Pb/Cu sandwich as a function of copper thickness. (Reproduced from Adkins and Kington [23].)

copper thickness much more rapidly for thin layers than for thick. As the copper thickness is increased

from zero, the value of $\Delta(x)$ at the interface is reduced and in addition, it decays across the copper film. Above some thickness of copper of the order of a coherence length (about 400 Å in this case), $\Delta(x)$ becomes more or less constant at the interface and only the decay across the copper is measured. These results are for a temperature of about 2.2 °K; in fact the temperature dependence of Δ_m was small below 4.2 °K.

From their results, Adkins and Kington deduce a value of -0.01 for $(NV)_{Cu}$, the possible limits being -0.10 and $+0.06$.

Qualitatively similar results have been obtained by other authors [26, 28, 29]. The most important deduction is the presence of an energy gap in the excitation spectrum at the copper surface which clearly demonstrates the existence of a proximity effect.

Finally, it should be mentioned that Rowell and McMillan have observed oscillations in the $I-V$ characteristic of a junction on the N side of a « clean » NS sandwich caused by electron interference in the normal metal [30]. A similar effect for low energy excitations ($< \Delta_m$) was predicted by de Gennes and Saint-James [44].

(ii) DIRTY SYSTEMS ($l \ll \xi$) NEAR T_{CNS} — Claeson et al. [24, 25] have performed experiments on the system Al/Al₂O₃/Pb/Ag, in which the lead was fairly thin so that the transition temperature of the sandwich was considerably lower than the bulk value. The samples were prepared at low temperatures so as to be in the dirty limit. They found that the $I-V$ characteristic was again smeared out, that is the density of states above the gap was heavily depressed and the conductivity at zero voltage was greatly enhanced by comparison with a B. C. S. superconductor. However, it should be remarked that this is really only an exaggerated version of what was observed by Adkins and Kington on the lead side of their comparatively clean Pb/Cu sandwiches at temperatures well below T_{CNS} . This emphasizes the difficulty of preparing specimens which really meet the demands of the theoretical models. The $I-V$ characteristics were in fact in reasonable agreement with the Fulde-Maki theory. For a given sandwich, the tunnelling characteristic behaved as if the gap in the lead were reduced significantly more than the transition temperature, just as in the experiments involving magnetic impurities. However, « quasi-gaplessness » was observed only very near T_{CNS} ; at lower temperatures a gap appeared. The characteristics obtained from tunnelling experiments on the silver side were not adequately explained in the Fulde-Maki theory.

Guyon et al [27], have undertaken similar experiments on In-Bi/Zn sandwiches, the In-Bi alloy having a

short mean free path. They obtained similar results for tunnelling on the S side and rather more detailed data for the N side. De Gennes and Mauro showed that near T_{CNS} the value of the pair potential on the surface of the normal metal [$\Delta(N)$] is given by :

$$\Delta^2(N) \propto 1 - (dI/dV)_0 \propto (1 - T/T_{\text{CNS}})^2. \quad (18)$$

Figure 14 shows the experimental results obtained for three thicknesses of normal metal deposited on a bulk lead film, compared with the predictions of the theory. We see that there is good agreement and in particular

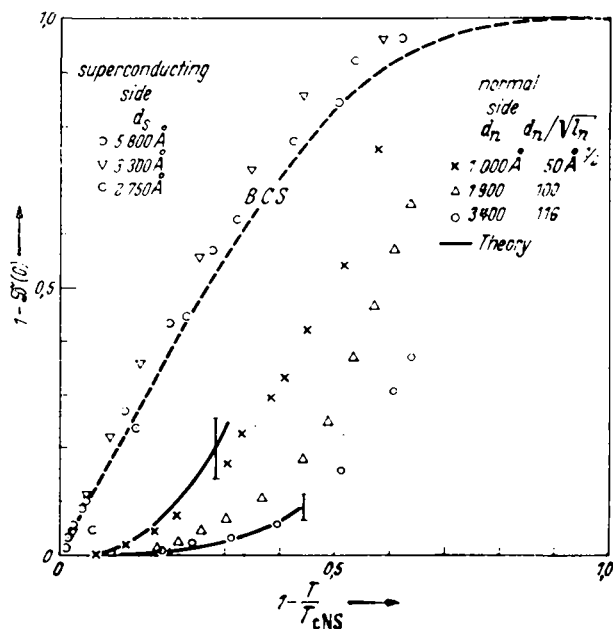


FIG. 14. — $(1 - (dI/dV)_0)$ against $(1 - T/T_{\text{CNS}})$ for tunnelling into a dirty NS sandwich : (1) the S side, when S is very thick, showing the comparison with the B. C. S. prediction (---); (2) the N side, for 3 different thicknesses, together with the theoretical prediction (---). (Reproduced from Guyon et al. [27].)

that there is a « quasi-gapless » region near T_{CNS} . At lower temperatures, a well-defined gap appears and the distinction between these experiments and those on the « clean » samples of (i) is then less obvious. For comparison, Figure 14 also shows the value of $(1 - (dI/dV)_0)$ obtained from tunnelling on the S side when the superconductor is very thick so that the presence of the normal metal does not affect the outer surface. The agreement with the B. C. S. prediction [37] is good : near T_{CS} , there is a linear dependence of $(1 - (dI/dV)_0)$ on $(1 - T/T_{\text{CS}})$ implying that Δ varies as $(1 - T/T_{\text{CS}})^{1/2}$. These results depict very clearly the difference between the intrinsic superconductivity of the lead and the induced superconductivity in the copper.

It would appear that near T_{CNS} there is a fair measure of agreement between the experiments and the theoretical predictions. However, an even better check on the theory could be obtained by examining specimens in which the film of superconductor is rather thin so that T_{CNS} is heavily depressed and the pair-breaking effect of the normal metal considerable; unfortunately, the preparation of such samples may be very difficult technically.

VII. The use of the proximity effect to estimate $(NV)_{\text{Cu}}$. — A number of authors have used the proximity effect to estimate NV in normal metals and their results for copper are summarized in Table I. It is clear

TABLE I

The values obtained by various authors for the interaction parameter NV and the corresponding transition temperature T_{CN} for copper.

	Method	$(NV)_{\text{Cu}}$	Estimated Limits	Corresponding T_{Cu} °K
Hilsch (1962)	T_{CNS} of NS	+ 0.05	+ 0.10 - 0.06	1.5×10^{-2} 7×10^{-7}
Hauser et al. (1964)	T_{CNS} of NS	+ 0.09		6×10^{-3}
Minnigerode (1966)	T_{CNS} of NS	+ 0.116		7×10^{-2}
Adkins and Kington (1966)	Tunnelling	- 0.01	+ 0.06 - 0.10	2×10^{-5}
Clarke	SNS		< 0.11	6×10^{-2}

that $(NV)_{\text{Cu}}$ is small, but its actual value very uncertain. The fairly large experimental uncertainties usually associated with thin films partly account for the discrepancies, but in addition there are difficulties concerning the precise definition of NV . Present theories [40, 42, 46, 53, 57, 65, 69] replace the retarded electron-electron interaction [48, 58] by an instantaneous interaction V whose Fourier transform is sharply cut off in momentum space. Jacobs [53] has shown that the value of $(NV)_{\text{Cu}}$ deduced from a given set of experimental data may be changed enormously by performing the cut-off in different ways. The theory is relatively insensitive to the value of $N_{\text{N}} V_{\text{N}}$ at temperatures well above the corresponding T_{CN} : this is illustrated by the results of Hauser and Theuerer for aluminium (Fig. 5) and can also be seen by inspection of equation

(8). Consequently relatively small errors in experiment or theory give rise to large discrepancies in $N_N V_N$. However, as we reduce the temperature, the dependence of the theory on T_{CN} increases; for example, if $T_{CN} = 10^{-2}$ °K, k_N (equation (8)) varies rapidly below about 0.3 °K. We thus have a technique which enables us to observe the *approach* to the superconducting state at temperatures well above the critical value. It is apparent that reliable data on $N_N V_N$ in metals where it is very small can be obtained only from experiments performed at very low temperatures.

VIII. Conclusions. — We have seen that there is reasonable agreement between theory and experiment for the transition temperature of NS sandwiches and the critical current of SNS sandwiches, which involve only the behaviour of the superfluid. On the other hand, more complicated calculations on the excitation spectra of NS sandwiches have received only partial verification by tunnelling experiments, which are themselves sometimes difficult to interpret. However, it is certainly true to say that the basic ideas of the proximity effect have been adequately confirmed by experiment.

Attempts to use the proximity effect to estimate a value of NV in metals where it is small have so far proved unsuccessful.

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DISCUSSION

Mr. BURGER. — Ne serait-il pas préférable de tester les théories d'effets de proximité entre deux métaux de points de transition très différents tels que Pb/W, plutôt qu'avec le système Pb/Cu ?

Mr. CLARKE. — I agree, although the preparation of the W films may present a problem.

Mr. DE GENNES. — Quels sont les effets de champ magnétique sur le courant critique des trilames Pb/Cu/Pb ?

Mr. CLARKE. — The supercurrent flowing through the junction penetrates into it a distance

$$\lambda_J = (\hbar c^2 / 8 \pi j_1 e d)^{1/2},$$

where λ is the penetration depth of the superconductor, $d = 2(a + \lambda)$ is the magnetic thickness of the barrier and j_1 the critical current density [54]. To discuss the effect of an external magnetic field we consider the two limiting situations in which λ_J is much greater than or much less than w , the width of the junction (i.e. of the lead strip). If $\lambda_J \gg w$, the current flows uniformly through the junction and the critical current is of the form $|\sin(\pi\Phi/\Phi_0)/(\pi\Phi/\Phi_0)|$, where Φ is the flux threading the junction and Φ_0 the flux quantum. This is what is usually observed in the case of SIS junctions. However, if we choose a junction in which j_1 is very large, we can obtain a situation where $\lambda_J \ll w$, that is, there is substantial self-field limiting. In this case, there is initially a linear decrease in critical current with increasing field until the thermodynamic critical field is reached (typically a few gauss). At this point, flux penetrates into the junction and a further increase in field produces the usual oscillations. The initial linear dependence of I_C upon H is simply the Meissner effect: a circulating current is set up in the junction to screen out the applied field and this has the effect of reducing the total supercurrent that the junction is able to sustain.